Environ Earth Sci (2015) 74:6481–6488
DOI 10.1007/s12665-015-4731-x

THEMATIC ISSUE

Thermodynamics study on the generation of electricity via CO_2-mineralization cell

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Received: 16 June 2015 / Accepted: 7 July 2015 / Published online: 23 August 2015

Abstract Carbon dioxide (CO_2) is the principal cause of the greenhouse effect which is due to the excessive consumption of fossil fuels. Several methods have been proposed for reducing CO_2 emissions in the atmosphere but none has been completely successful. Recently, a novel technique called the CO_2-mineralization cell (CMC) technique, based on CO_2 mineralization and utilization (CMU), was proposed by our group. This converts chemical energy from the CO_2 mineralization reaction into electricity while also producing highly valuable chemical products. However, some confusion and doubts still exist about its theoretical feasibility. Herein, a thermodynamics study and analysis of the CMC were conducted and the feasibility of generating electricity by CO_2-mineralization reaction confirmed theoretically. The corresponding theoretical electromotive force and the maximum electric energy production per 1t carbon dioxide completely consumed under standard conditions were also calculated, and their influential factors fully discussed.

Keywords CO_2-mineralization cell · Thermodynamics study · Generation of electricity · Theoretical electromotive force · Maximum electric energy production

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Introduction

Global atmospheric carbon dioxide levels are increasing sharply with the excessive use of fossil fuels and aggravate the greenhouse effect, which is a serious threat to the human environment (Mikkelsen et al. 2010; Xie 2010a). Data produced by the International Panel on Climate Change (IPCC) indicate that the content of CO_2 in the atmosphere could increase to 570 ppm, resulting in a temperature rise of 1.9 °C around the world by the end of twenty-first century (Yang et al. 2008). At the same time, the growing demand for energy is another global issue as yet unresolved. Hence, it is essential to develop new forms of energy before the fossil fuels are depleted.

As is well known, there are several methods of reducing CO_2 emissions in the atmosphere, such as the CO_2 capture and storage (CCS) technology or by utilizing CO_2 as a chemical raw material (Aresta 2010; Barrow 1973; Hou et al. 2014; Klotz and Rosenberg 1950; Liu et al. 2014; Sheng et al. 2015; Xie et al. 2014a, 2015a; Zhang et al. 2015b). To mitigate the adverse effect on the environment and ensure the sustainable use of fossil energy, numerous studies have been carried out to sequestrate CO_2 underground (Kolditz et al. 2015; Zhang et al. 2015a). The so-called geological sequestration technology has been researched and trialed globally as a way of storing carbon (Hou et al. 2015). The feasibility of this solution has,

especially, been the focus of a detailed research project by a European group under conditions close to those beneath the North Sea (Holloway et al. 1996). As reported recently, CO_2 has also been injected into deep un-minable coal seams both as a means of storage and to increase the exploitation of methane using a method similar to that for the enhanced oil recovery (EOR) programs in the Texas oilfields in the United States (Hu et al. 2010; Pearce et al. 1996). Although carbon dioxide can be sequestered in porous reservoir rocks below a certain depth using the present technology, the problems of storing it in the subsurface cannot be resolved fundamentally due to the possibility of long-term geological disasters when it leaks to the surface.

It is well known that the mineralization of CO_2 is a chemical process between carbon dioxide and alkaline minerals to generate carbonated compounds which are stable under natural conditions (Seifritz 1990; Xie et al. 2013). To date, several experimental and theoretical studies on carbonation reactions have been carried out under different conditions using a variety of natural minerals (e.g., serpentine, forsterite and wollastonite) in an attempt to reduce CO_2 levels in the atmosphere (Lackner et al. 1995). However, direct experimental carbonation processes are usually extremely slow and difficult under room temperature, as the carbonate generated will decompose when the temperature is too high. Therefore, the traditional carbonation process is not a satisfactory solution to carbon dioxide problems.

Although carbon dioxide exists stably as an inert gas in the atmosphere, under certain conditions it can also react with specific alkaline compounds in a few minutes or hours in a process called accelerated carbonation (Pan et al. 2012). During this progress, carbon dioxide may interact with an alkaline material, including natural silicate-minerals and industrial residues, to form a stable carbonate in a short time, under specific temperature and humidity conditions. Alkaline solid wastes have been proved to be effective raw materials to react with CO_2 (Nyambura et al. 2011; Wang and Yan 2010). However, the necessary conditions for these reactions are always harsh and the loss outweighs the gain from an economic point of view, as the process would consume additional energy and generate more CO_2 emissions.

Meanwhile, a new method known as CO_2 mineralization and utilization (CMU) has been proposed by our research group to resolve CO_2 problems. Under this technique, the captured CO_2 is utilized as a raw material to manufacture high value carbonation products from both an economic and an environmental point of view (Xie 2010b; Xie et al. 2012, 2013, 2015b, c). During this process, carbon dioxide is carbonated by natural minerals and solid waste under conditions of low energy consumption and low cost. Unlike the CCS method, this is an effective way to reduce carbon dioxide emissions while also creating economic value.

A novel technique to generate electricity using a CO_2-mineralization cell (CMC) was reported recently (Xie et al. 2014c), in which portlandite [$Ca(OH)_2$] and CO_2 are the raw materials. H_2 gas is catalytically used up at one electrode and regenerated at the second electrode to introduce electrons or an H_2-induced current in the external circuit of the cell. This design allows a stable current and electric energy to be generated by the CMC cell. However, some doubt still exists about its theoretical feasibility to be a revolutionary method to deal with carbon dioxide, especially as the mechanism for the process of electricity generation via carbonation is currently not well known. At the same time, the corresponding electromotive force and the maximum electric energy produced under different conditions are also unknown. In this paper, the thermodynamics of generating electricity by CO_2 mineralization has been studied, and a theoretical analysis and feasibility of the relevant reactions conducted. In addition, the corresponding theoretical electromotive force and the maximum electric energy produced at standard conditions were theoretically calculated and the influential factors discussed.

The chemical thermodynamics of the generation of electricity using the CMC system

Chemical thermodynamics is the interrelation between work and heat which may change after physical changes of state or chemical reactions, and must obey the three laws of thermodynamics. Thermodynamic functions [e.g., internal energy (U), enthalpy (H), entropy (S), Gibbs free energy (G)] are obtained by experience and verified by rigorous logical reasoning, and only involve the beginning and end of the change in state regardless of the process. Chemical thermodynamics can only solve problems of a system in a state of equilibrium (Klotz and Rosenberg 1950; Ott et al. 2001).

According to the second law of thermodynamics, the stability of a system under certain conditions can be described by the change in the Gibbs free energy, ΔG of the corresponding system. The free energy G is a function of pressure, temperature and the material quantity of each component in the system. It is given as follows:

$G = f(T, p, n_1, n_2 \ldots n_m)$, where m represents the number of components in the system and hence

$$\Delta G = \left(\frac{\partial G}{\partial T}\right)_{p,n_m} \mathrm{d}T + \left(\frac{\partial G}{\partial p}\right)_{T,n_m} \mathrm{d}p + \sum_{i=1}^{m} \left(\frac{\partial G}{\partial n_i}\right)_{T,p,n_j} \mathrm{d}n_j \quad (1)$$

Under a certain temperature T and pressure P,

$$G = \sum_{i=1}^{m} \mu_i n_i \tag{2}$$

where μ_i represents the chemical potentials of each component; n_i represents the amount of substance of each component.

The change in Gibbs free energy, ΔG, is

$$\Delta G = \mathrm{d}G_{T,p} = \sum_{i=1}^{m} \mu_i \mathrm{d}n_i \tag{3}$$

The progress of a chemical reaction can be predicted based on the change in Gibbs free energy, i.e., when $\Delta G < 0$, the reaction process can occur spontaneously; $\Delta G = 0$ implies that the system is in a state of equilibrium and therefore stable, while a system with $\Delta G > 0$ will not participate in any reaction.

Gibbs free energy is a measure of the amount of useful work that could be gained from a thermodynamic system under constant temperature and pressure conditions. It is a significant guide to the direction and results of the experiment (Klotz and Rosenberg 1950; Ott et al. 2001). In a balanced, reversible process for a reaction occurring under conditions of constant temperature and pressure, the change in Gibbs free energy ($\Delta_r G_m$) measures the maximum non-volume work ($-W_{f,\,\max}$) obtainable from the system, i.e.,

$$\Delta_r G_m = -W_{f,\max} \tag{4}$$

When the above equation is applied to a reversible electrochemical CMC cell, the non-volume work of the system will be equivalent to the electrical work, which is equal to the numerical product of the electromotive force E and the electric constant F, i.e.,

$$\Delta_r G_m = -ZFE \tag{5}$$

where E represents the reversible electromotive force of the cell, Z represents the measurement coefficient, i.e., the number of the electrons released at the negative electrode, and F represents the Faraday constant.

At the standard conditions of 298.15 K and 1 atm, E^θ is

$$E^\theta = -\frac{\Delta_r G_m^\theta}{ZF} \tag{6}$$

According to the above formula, the electromotive force E of a cell's reaction and the maximum electric energy produced $\Delta_r G_m$ can be calculated under certain conditions.

Though carbon dioxide is hard to deal with, it is always less stable than the carbonated salt. From the point of view of thermodynamics, the standard Gibbs free energy of the formation of the carbonated salts is about 60–180 kJ/mol lower than that of carbon dioxide which implies that carbonation could be a satisfactory solution for dealing with carbon dioxide (Wendt et al. 1998). Although the change in Gibbs free energy is far less than 0, the energy of this chemical reaction cannot be converted directly into electricity as the reaction is non-redox, without any transfer of the electrons. In our previous paper (Xie et al. 2014c), a CMC system was proposed that would obtain automatically. In this study, however, an electrocatalyst and H_2 have been introduced to the system. At the anode surface coated by an electrocatalyst, such as Pt, H_2 will give up electrons to the electrode leaving the protons to react quickly with the abundant hydroxyl ions to form water. When the two electrodes are joined by a conducting wire, these electrons will flow in the external circuit to the cathode to react with the protons produced by the CO_2 in the aqueous solution to regenerate H_2 at the cathode surface. The flowing electrons will generate a current called H_2-induced current and the energy from the acid–base neutralization reaction will be released as electrical energy. If carbon dioxide and the alkaline solution are mixed together, they will interact with each other releasing chemical energy in the form of heat. In this case, a cation ion-exchange membrane is placed between the two pole electrodes, which prevents a chemical reaction process but allows the ion diffusion process to take place. The corresponding reaction mechanisms are also illustrated in the electromotive CMC cell (Fig. 1).

With a H_2-induced current and an ion-exchange membrane separating the reactants, the complex system of carbonation reaction generating electricity can be considered as a special reversible cell. Using thermodynamics, the corresponding electromotive force and the maximum electric energy produced by the cell can be predicted for a certain temperature and pressure.

Taking sodium hydroxide and carbon dioxide as the raw materials, the total complex reaction equation can be written as shown below:

$$\mathrm{CO_2(g)} + \mathrm{NaOH(aq)} \xrightarrow[\mathrm{CMC}]{} \mathrm{NaHCO_3(aq)} + \text{Electric energy} \tag{7}$$

The standard Gibbs free energy of formation ($\Delta_f G_m^\theta$) of the reactants and products in the above complex reaction equation and their corresponding standard molar Gibbs free energy of reaction ($\Delta_r G_m^\theta$) are listed in Table 1 (McGlashan 1973).

It is well known that $\Delta_r G_m^\theta$ is the Gibbs free energy change of the reaction when 1 mol of components reacts at the standard conditions of 298.15 K and 1 atm, while the $\Delta_r G_m$ is the real criterion for the reaction under constant temperature and pressure considering that the components are not reacting under standard conditions. The relationship between them can be expressed as below (Ott et al. 2001):

$$\Delta_r G_m^\theta = \Delta_r G_m - RT \ln Q \tag{8}$$

where Q is the activity product at certain temperature and pressure.

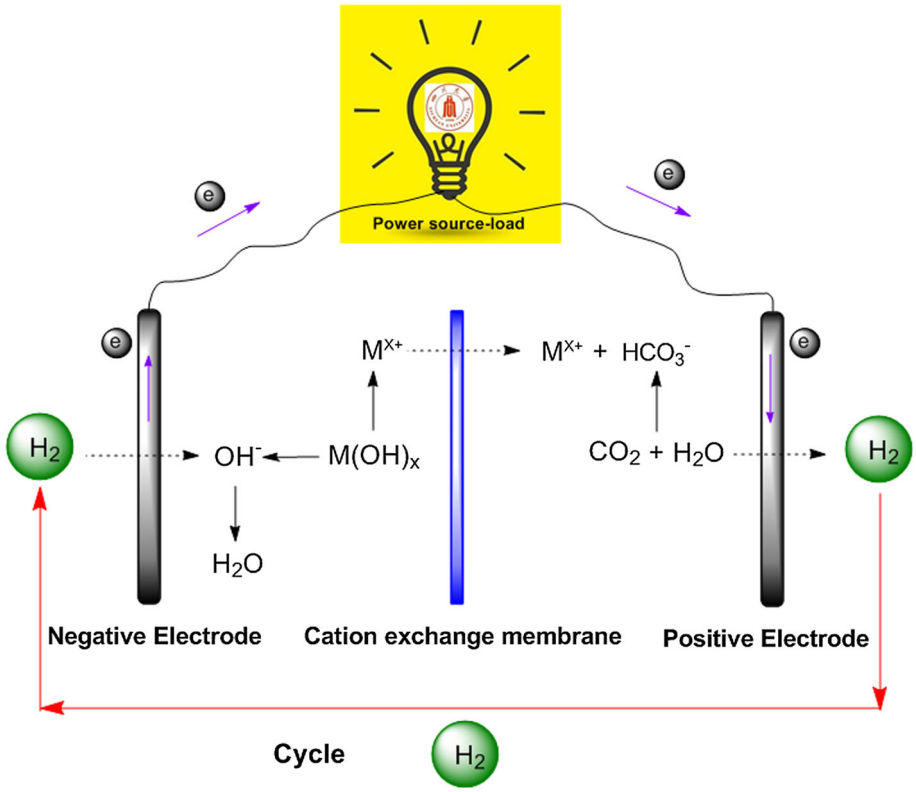

Fig. 1 A schematic diagram of a CO_2-mineralization cell, a system in which electricity is generated. $M(OH)_X$ represents the alkaline feedstock (e.g., sodium hydroxide and calcium hydroxide) and M^{X+} represents its corresponding cation. The *purple and red arrows* represent the directions of the electron transfer and hydrogen circulation, respectively

Table 1 $\Delta_f G_m^\theta$ of the reactants and products and $\Delta_r G_m^\theta$ of the total reaction of NaOH and CO_2

	Materials	$\Delta_f G_m^\theta$/kJ mol^{-1}	$\Delta_r G_m^\theta$/kJ mol^{-1}
Reactants	CO_2 (g)	−394.38	−35.41
	NaOH (aq)	−419.15	
Products	$NaHCO_3$ (aq)	−848.94	
Reaction	$CO_2(g) + NaOH(aq) \xrightarrow[CMC]{} NaHCO_3(aq)$ + Electric energy		

$$\prod_B a_B^{\upsilon_B} = Q \tag{9}$$

where υ_B represents the stoichiometric number of component B, a_B represents the activity of component B, and Q is the activity product at certain temperature and pressure; the relative activity of the gas can be expressed as $a_B = \frac{f_B}{p^\theta}$($f_B$ represents the fugacity coefficient of gas component B and p^θ represents the standard gas partial pressure).

Combined with the Eq. (7), the $\Delta_r G_m$ can be expressed as shown in formula (10):

$$\Delta_{r_1} G_m^\theta = \Delta_{r_1} G_m - RT \ln Q_1 = \Delta_{r_1} G_m - RT \ln \frac{a_{HCO_3^-}}{\frac{p_{CO_2}}{p^\theta} * a_{OH^-}} \tag{10}$$

Under the conditions of 298.15 K and 0.1 MPa, the concentration of the bicarbonate ions is calculated as 1.2×10^{-4} mol/L when the dissolved carbon dioxide is saturated. The change in Gibbs free energy of the reaction is calculated as −57.79 kJ/mol with 1 mol/L sodium hydroxide. Altogether the complex carbonation system will release 57.79 kJ energy in the form of electricity and heat when 1 mol CO_2 is consumed by the reaction. According to the expression in Eq. (5), i.e. $\Delta_r G_m = -ZFE$, and the reaction Eq. (7), in which the value of Z is one, then the corresponding theoretical electromotive force E can reach up to 0.599 V. Therefore, the system can theoretically generate 364.84 kW h electric energy when 1t carbon dioxide is completely mineralized.

When the option of alkaline feedstock is considered, portlandite will play an important role in the carbonation reactions. Similar to sodium hydroxide, the total complex reaction equation is as shown below:

$$2CO_2(g) + Ca(OH)_2(s) \xrightarrow[CMC]{} Ca(HCO_3)_2(aq) + \text{Electric energy} \tag{11}$$

Similarly, the standard Gibbs free energy of the formation ($\Delta_f G_m^\theta$) of the reactants and products of the complex reaction Eq. (11) and the corresponding standard molar

Gibbs free energy of reaction ($\Delta_r G_m^\theta$) are displayed as Table 2 (Barrow 1973).

The corresponding $\Delta_r G_m$ can be expressed as the formula (12):

$$\Delta_{r_2} G_m^\theta = \Delta_{r_2} G_m - RT \ln Q_2 = \Delta_{r_2} G_m - RT \ln \frac{a^2_{HCO_3^-} * a_{Ca^{2+}}}{p^2_{CO_2}/p^{\theta^2}} \quad (12)$$

Under the conditions of 298.15 K and 0.1 MPa, when the dissolved carbon dioxide is saturated and the portlandite is in excess, the concentration of the calcium and bicarbonate ions is calculated as 1.1×10^{-2} mol/L and 1.2×10^{-4} mol/L, respectively (Barrow 1973; Linke and Seidell 1958). The change in Gibbs free energy of reaction is calculated as −96.55 kJ/mol. This result implies that the complex carbonation reaction will in this case release up to 96.55 kJ energy in the form of electricity and heat when 2 mol of CO_2 is consumed by the reaction. A little different from that of sodium hydroxide, the measurement coefficient of the electrons in the electrode reaction is 2, so the corresponding theoretical electromotive force can reach a maximum value of 0.500 V. Therefore, this system can theoretically generate up to a maximum of 304.77 kW h of electricity when 1t carbon dioxide is completely used up (mineralized).

However, the calcium bicarbonate generated will transform into solid calcium carbonate adsorbed or deposited onto the surface of the anode pole blocking the active reaction site during the reaction progress. Taking the kinetic factor into consideration, soluble sodium salts are added to the cathode solution so that the carbonated products remain in the form of sodium bicarbonate instead of calcium carbonate. Sodium carbonate and sodium sulfate, in particular, can be good additives for this carbonation system. The corresponding reaction equations and thermodynamic data for this system are shown in Table 3.

The related reaction equations and thermodynamic data of other complex reactions with different available alkaline feedstock are shown in Table 4.

Influential factors on the generation of electricity via CMC system

In the above section, the equilibrium electromotive forces of carbonation reactions through different pathways were discussed, confirming the possibility of electric power generation in theory and its mutual authentication with the experimental phenomena (Xie et al. 2014c). With regard to Eq. (11), the theoretical electromotive force is 0.5 V and the experimentally measured result is 0.452 V, which is reasonable considering that there was some overpotential loss at the electrodes. To improve the practice electromotive force, the influential factors will be analyzed as follows.

It is well known that Gibbs free energy of chemical reaction will be mostly affected by the temperature and concentration of both the reactants and products. As shown above, Gibbs free energy is closely related with the activity product of each component at a certain temperature and pressure.

According to expression in Eqs. (5) and (8), the electromotive force of a certain reaction is influenced by the activity product of each component at constant temperature and pressure (all reaction solutions are considered to be ideal liquids).

$$E = -\frac{\Delta_r G_m}{ZF} = E^\theta - \frac{RT \ln Q}{ZF} \quad (13)$$

As a result, in the course of these experiments, the controlled dosage ratio of the raw materials can increase the change in Gibbs free energy, thus improving the electromotive force theoretically and experimentally. For example, if sodium hydroxide and carbon dioxide are the raw materials, the change in Gibbs free energy of this reaction is expressed in the formula (10). When excess sodium hydroxide is added to saturation, the activity of the hydroxide ion will increase and the $\Delta_r G_m$ will reduce. The value of $\Delta_r G_m$ can reduced to −66.0 kJ/mol approximately and the theoretical electromotive force to about 0.684 V. For the same reason, the corresponding theoretical electromotive force will be higher if the partial pressure of the carbon dioxide is enhanced. A plot of the theoretical relationship between the concentration (of sodium hydroxide) and the ensuing electromotive force under standard state conditions is shown in Fig. 2 (Barrow 1973; Klotz and Rosenberg 1950; McGlashan 1973).

In the above calculation and analysis, all reaction solutions are taken as ideal liquids under standard conditions. However, the activity of each compound and the fugacity of carbon dioxide will always change at different temperatures or pressures. Similarly, the solubility of the

Table 2 $\Delta_f G_m^\theta$ of the reactants and products and $\Delta_r G_m^\theta$ of the total reaction of $Ca(OH)_2$ and CO_2

	Materials	$\Delta_f G_m^\theta$/kJ mol^{-1}	$\Delta_r G_m^\theta$/kJ mol^{-1}
Reactants	CO_2 (g)	−394.38	−41.64
	$Ca(OH)_2$ (s)	−896.76	
Products	$Ca(HCO_3)_2$ (aq)	−1727.16	
Reaction	$2CO_2(g) + Ca(OH)_2(s) \xrightarrow[CMC]{} Ca(HCO_3)_2(aq)$ + Electric energy		

Table 3 Reaction equations including the additives, sodium carbonate and sodium sulfate and the change in Gibbs free energy of reaction under the standard state conditions of 298.15 K and 0.1 MPa; E represents the corresponding theoretical electromotive force; Maximum generation represents the potential peak output when 1t carbon dioxide is completely mineralized (Barrow 1973; Linke and Seidell 1958)

Additives	$\Delta_r G_m^\theta$/kJ mol^{-1}	E/V	Maximum electric energy production per 1t CO_2 consumed/kW h
Sodium sulfate	−66.91	0.582	353.54
Sodium carbonate	−89.26	0.682	415.56
Reaction 1	$2CO_2(g) + Ca(OH)_2(s) + Na_2SO_4(aq) \xrightarrow[CMC]{} 2NaHCO_3(aq) + CaSO_4(s) + \text{Electric energy}$		
Reaction 2	$2CO_2(g) + Ca(OH)_2(s) + Na_2CO_3(aq) \xrightarrow[CMC]{} 2NaHCO_3(aq) + CaCO_3(s) + \text{Electric energy}$		

Table 4 Reaction equations for a variety of alkaline feedstock

Alkaline feedstock	$\Delta_r G_m^\theta$/kJ mol^{-1}	E/V	Maximum electric energy production per 1t CO_2 consumed/kW h
KOH	−35.41	0.599	364.84
$Mg(OH)_2$	−7.63	0.351	213.54
NH_3	−18.36	0.562	342.68
LiOH	−35.41	0.599	364.84
Na_2CO_3	−14.45	0.614	373.80
	Reaction equations		
KOH	$CO_2(g) + KOH(aq) \xrightarrow[CMC]{} KHCO_3(aq) + \text{Electric energy}$		
LiOH	$CO_2(g) + LiOH(aq) \xrightarrow[CMC]{} LiHCO_3(aq) + \text{Electric energy}$		
NH_3	$CO_2(g) + NH_3(g) + H_2O \xrightarrow[CMC]{} NH_4HCO_3(aq) + \text{Electric energy}$		
$Mg(OH)_2$	$2CO_2(g) + Mg(OH)_2(s) \xrightarrow[CMC]{} Mg(HCO_3)_2(aq) + \text{Electric energy}$		
Na_2CO_3	$CO_2(g) + Na_2CO_3(aq) + H_2O \xrightarrow[CMC]{} 2NaHCO_3(aq) + \text{Electric energy}$		

The values for the change in Gibbs free energy are calculated for the standard state conditions of 298.15 K and 0.1 MPa; E represents the corresponding theoretical electromotive force; Maximum electric energy generation represents the potential peak output when 1t carbon dioxide is completely mineralized; All the concentrations of the alkaline raw materials are considered to be 1 M, except $Mg(OH)_2$, and the dissolved carbon dioxide is saturated (Barrow 1973; Linke and Seidell 1958)

reactants also changes with temperature, for instance the solubility of sodium sulfate can range over one order of magnitude giving rise to an upheaval in the experimental results. Meanwhile, the value of $\Delta_r G_m$ varies with the temperature. As a result, choosing appropriate parameters under the prevailing environmental conditions will be very important for the cell's output power. However, once the generated current passes through the electrode, the Galvani potential difference will differ from the Nernst equation. The magnitude of the difference is called the overpotential (η). η reflects the degree of polarization of the electrodes which includes activation polarization, ohmic polarization and concentration polarization (Bard and Faulkner 1980). The relationship between current density and overpotential can be described by the Butler–Volmer Eq. (14):

$$j = j_0\left(e^{\frac{\alpha ZF}{RT}\eta} - e^{-\frac{\beta ZF}{RT}\eta}\right) \tag{14}$$

where η is overpotential, j is the current density, j_0 is the exchange current density, and α and β are the transfer coefficients.

The loss in reaction rate is the major factor for voltage loss at low current density. It can be realized from the B-V equation that the exchange current density is very important (i_0). When the j_0 is higher, the η is lower. There are several strategies for increasing the j_0 of the electrochemical reaction on the electrode: (1) Increase cell temperature. The j_0 of oxygen reduction reaction will increase 100 times when the temperature increase to 800 °C. (2) Use a more effective catalyst. The catalyst affects both the activation energy and the transfer coefficient. (3) Improve the

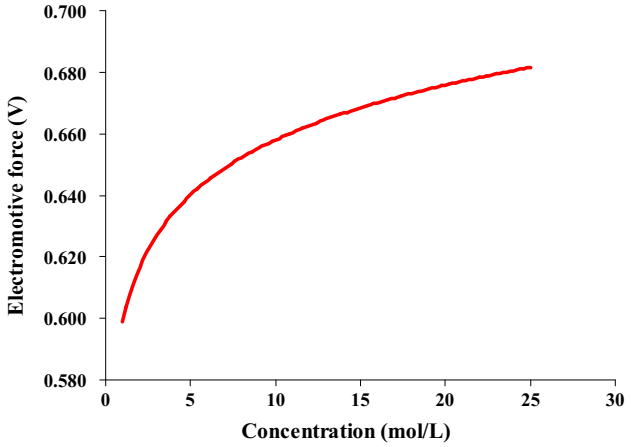

Fig. 2 Theoretical relationship between concentration (sodium hydroxide) and the resulting electromotive force (298.15 K and 0.1 MPa)

electrode roughness to increase the electrochemical active surface area. (4) Increase the gas pressure to increase contact between the reaction and the catalyst.

Conclusion and outlook

In this paper, the chemical thermodynamics of the corresponding carbonation reaction has been studied based on the experimental results of generating electricity via CO_2 mineralization. The thermodynamic feasibility of a CO_2 mineralization cell is discussed and demonstrated, providing the theoretical support for the experimental results.

Results of the thermodynamics calculation show that the theoretical electromotive force generated can be up to 0.684 V and the maximum electric energy generated when 1t carbon dioxide is completely carbonated can reach a maximum performance of 416.67 kW h under the standard state conditions of 298.15 K, 0.1 MPa and a saturated sodium hydroxide solution as the alkaline feedstock. Moreover, the influential factors for the actual cell potential loss were analyzed and resolving strategies proposed. These results can be an effective guidance for improving future experiments.

In the follow-up work, we shall study the kinetics of carbonation reactions generating electric power which may be an important guiding significance to the experimental process. We also intend to investigate different alkaline materials, including alkaline nepheline and organic amine compounds, used as feedstocks carried out from both a theoretical and an experimental point of view. In addition, different reaction conditions are being studied to attain the generation of a higher electromotive force and hence a larger electric power output.

Acknowledgments This work was financially supported by Natural Science Foundation of China (51254002 and NSFC 21336004) and the Ministry of Science and Technology (State Key Research Plan 2013BAC12B03).

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